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DECOMPOSITION AND INTERACTION OF PROPELLANT INGREDIENTS.(U)
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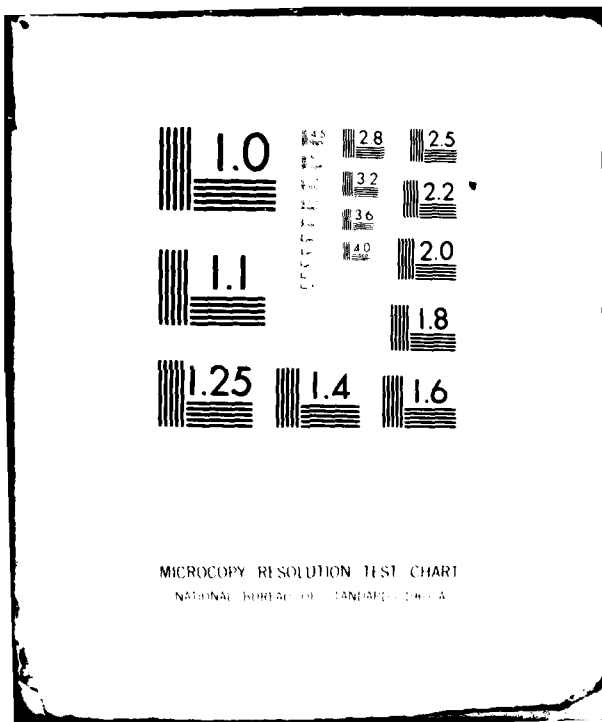
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NAVSEA Propulsion Research Program

Program Element 61153N

Decomposition and Interaction of
Propellant Ingredients.

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Technical Objective:

Investigate decomposition, deflagration, and interaction of conventional and new propellant ingredients with the goal of improving performance and safety, reducing cost to achieve specified performance, and providing methods to assure reproducibility during production and usage.

Background:

There is a continuous need in the field of solid propellant combustion to understand and solve problems such as cook-off hazards, burn rate anomalies, burn rate tailoring and control, combustion stability, combustion efficiency, and the use of high energy or highly aluminized propellants. The research effort of this program supplies the knowledge and fundamental data necessary to understand and to solve current and future combustion problems. It also generates the basis for the control of propellant combustion behavior, the development of new propellant systems, the use of novel ingredients, and the quality control of ingredients. The research data and results furthermore are indispensable for consultation in the combustion community, for the achievement of specific combustion goals, and for a critical assessment and application of data reported in the literature.

Approach:

The program utilizes a wide variety of experimental methods as well as theoretical studies in specific areas such as burn rate modeling. These methods have been well documented in numerous presentations, publications, and patents. They allow the study of catalyzed and uncatalyzed decomposition of propellant oxidizers, binders, and metal fuels, and the evaluation of combustion characteristics, combustion performance, and combustion stability of experimental and production lot propellants. Where required, established test methods are modified or new methods are developed to meet specific needs such as in particle size analysis or ingredient modification. In-house or commercial analytical capabilities are employed whenever feasible or promising. The interpretation of results is facilitated by limiting the number of sample and test parameters as much as possible and by rigorous control of test variables. In the process, new options for tailoring of propellant combustion behavior, new propellant systems, and quality control of ingredients are involved.

Progress:

The FY 80 effort comprised the investigation of HMX decomposition,¹ HMX combustion,² and the burning behavior of TATB.³ The application of results achieved under this program in the area of aluminum combustion was discussed in two presentations.^{4,5}

Decomposition of HMX

Propellants containing HMX are the object of considerable research and development efforts because of advantages gained by the use of this particular ingredient. Problems encountered by the replacement of more conventional oxidizers by HMX, the complexities of HMX decomposition and deflagration, and the desire to fully utilize the potential of HMX in propellant formulations prompted increased research in recent years concerning the decomposition and combustion behavior of HMX.

Several decomposition mechanisms have been proposed as a result of past investigations, including a number of kinetic parameters. During FY 80, HMX was studied to gain additional details of its thermal decomposition in the solid state and, most important, to obtain such data which would enhance the search for effective reaction rate modifiers. Some qualitative work with liquid phase HMX was also performed.

In the solid phase at atmospheric pressure, HMX decomposed as shown in Figure 1 in the temperature range 229-269°C. An acceleratory period which extends to a fractional decomposition of approximately 20% is followed by a decay period. The production of a rather stable residue of a few weight percent is shown in Figure 2 suggesting that the experimental weight loss curves fall below expected values near 100% decomposition. The increase of sample size also increased the decomposition rate (Figure 3). Rather surprising was the sensitivity of decomposition rate on gaseous environment as shown in Figure 4 for the isothermal decomposition at 259°C. Erofeev's equation was used to evaluate the rate constants for a first order reaction during the decay period. Figure 5 shows the temperature dependence of the decomposition rates in air and oxygen. The constancy of the activation energy suggests that environmental gases influence the concentration of certain species which participate in the decomposition rather than the mechanism of the reaction. The effectiveness of oxygen as a decomposition retardant can be explained by dissociation of NO_2 from the HMX molecule and the influence of oxygen on the equilibrium of thermal NO_2 dissociation. Consequently, compounds which react readily with the mixture of NO_2 and its dissociation products, would have a strong effect on HMX decomposition. One gas and several compounds have been used successfully to change the decomposition rate of HMX. It was found that in order to be an effective rate modifier, the additive should (a) produce species that will chemically react with the decomposition products of HMX and (b) additive as well as HMX should generate reactive species in the same temperature range.

The morphology of HMX decomposition in the solid state was examined by scanning electron microscopy. A photographic presentation of details can be found in reference (1).

The appearance of liquid phase during heating of HMX could not be marked by a melting point. At constant sample size, the time necessary to liquefy HMX in the temperature range 266-276°C increased sharply with decreasing temperature (the activation energy for the process would be

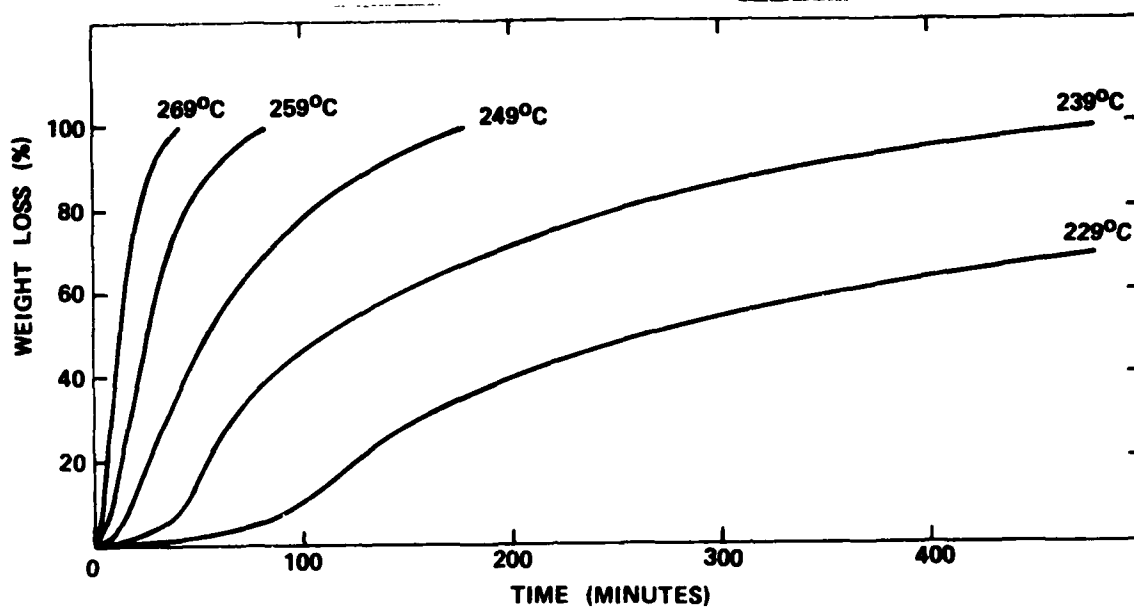


Figure 1. Weight Loss as Function of Time for HMX Decomposition in 93 kPa of Air

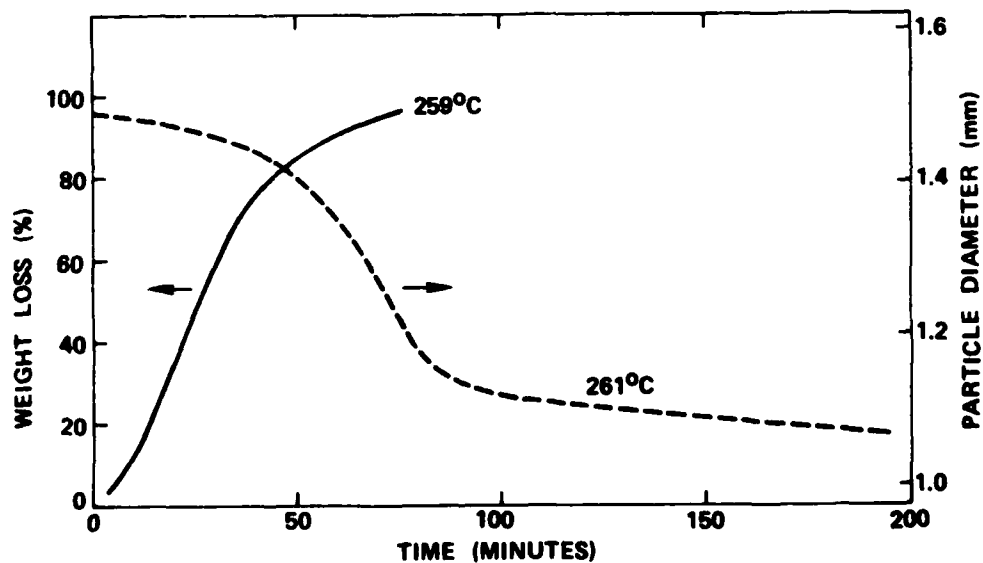


Figure 2. Weight Loss and Particle Size as Function of Time for HMX Decomposition in 93 kPa of Air

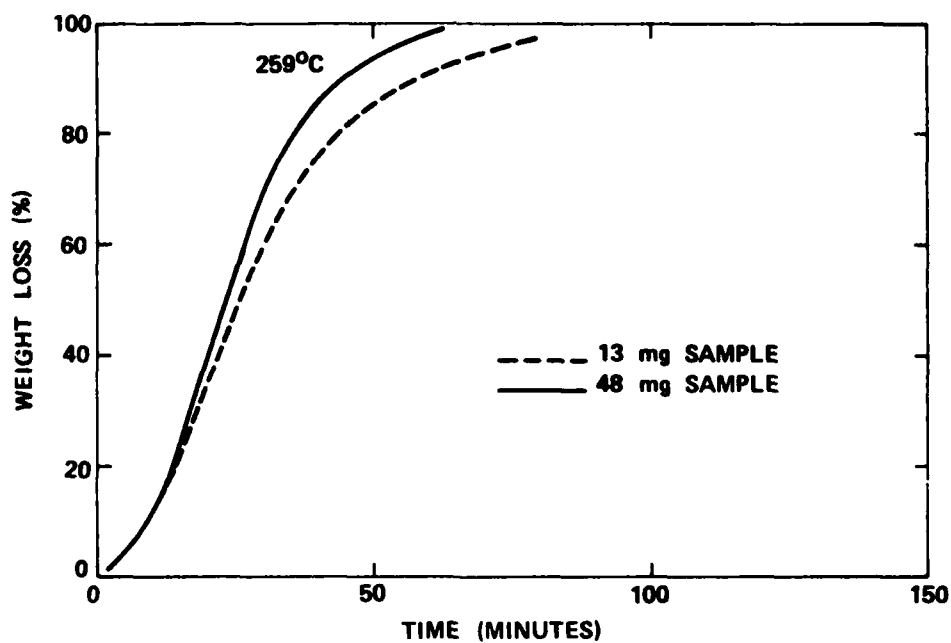


Figure 3. Effect of Sample Size on HMX Decomposition at 259°C in 93 kPa of Air

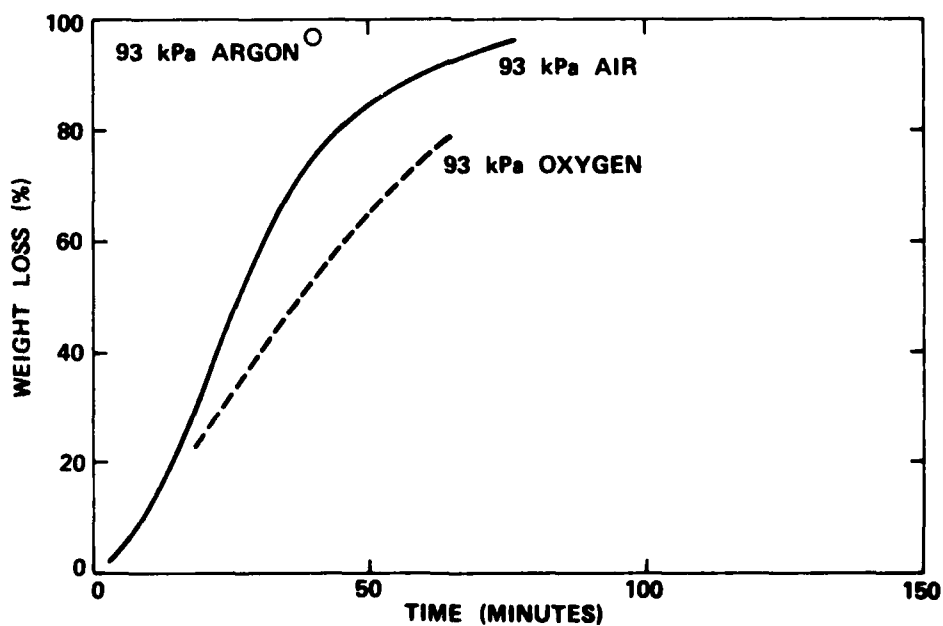


Figure 4. Effect of Gaseous Environment on HMX Decomposition at 259°C. Single point (O) was obtained for 40 minutes decomposition in argon

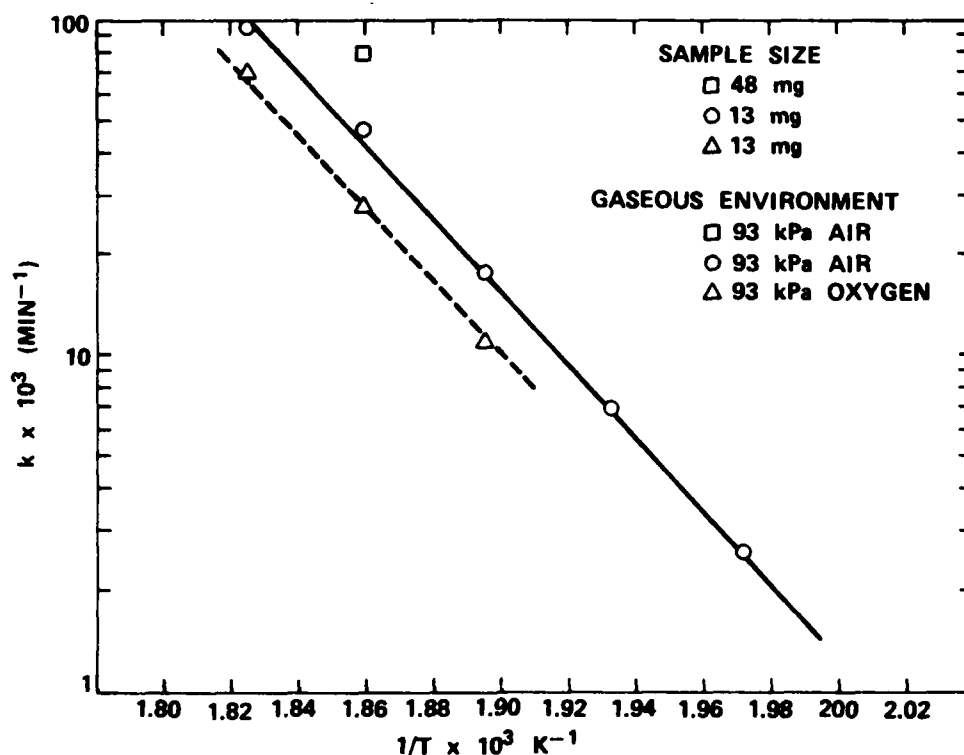


Figure 5. Temperature Dependence of Rate Constants for HMX Decomposition in 93 kPa of Air (—) and Oxygen (----). Single point (\square) was obtained with 48 mg samples

approximately 210 kcal/mole according to preliminary results). The low temperature of liquefaction depended on particle size and concentration (autocatalysis and/or self-heating) and may also depend on competition with other decomposition reactions.

Combustion of HMX

At high pressures ($p > 1000$ psi) considerable disagreement exists between investigators with respect to the burn rate of HMX. In particular, there is some uncertainty with regard to the presence of a transition to a high slope region in the burn rate versus pressure curve. A region of increased slope was indeed found between 7,500–20,000 psi. Arguments for sample break-up and/or convective burning as cause for slope increase were presented earlier.^{6,7} In view of the uncertainties, closed bomb combustion tests were conducted with carefully sieved, high purity HMX powder.² The measured burn rates are shown in Figure 6 as a function of pressure. No significant slope breaks of the burning rate were found. At high pressures the burn rate exponent was unity. For pressures up to 7,000 psi, gas phase kinetics were found to be an important consideration, especially for finely divided HMX.

Burning Behavior of TATB

Because of its excellent thermal stability and resistance to impact initiation, TATB has been suggested as an alternate ingredient or substitute for HMX in explosives and high energy propellants. The burn rate of TATB as a function of pressure was measured in order to assess the potential of TATB as an alternate or substitute of HMX. The self-deflagration rate of TATB was found to be an order of magnitude lower than that of HMX over the range 2,000-15,000 psi (Figure 6). Also, TATB did not sustain self-deflagration at low pressures ($p < 1,500$ psi) in the sample configuration and apparatus used.

Survey of Nitramine Decomposition and Combustion

A survey of the decomposition and combustion of HMX and RDX and of propellants incorporating these ingredients has been made and will be reported as a chapter in a forthcoming American Institute of Aeronautics and Astronautics Progress Series volume.

It is planned to complete the isothermal decomposition tests (see Section A) in FY 80 and to start or continue with the analysis of reaction products and a more detailed investigation of liquid HMX respectively. The investigation of additives for reaction and burn rate modification will include AP. AP is particularly interesting because it generates NH_3 and HClO_4 , which both can react with HMX. Burn rate studies will be expanded to include HMX/AP mixtures.

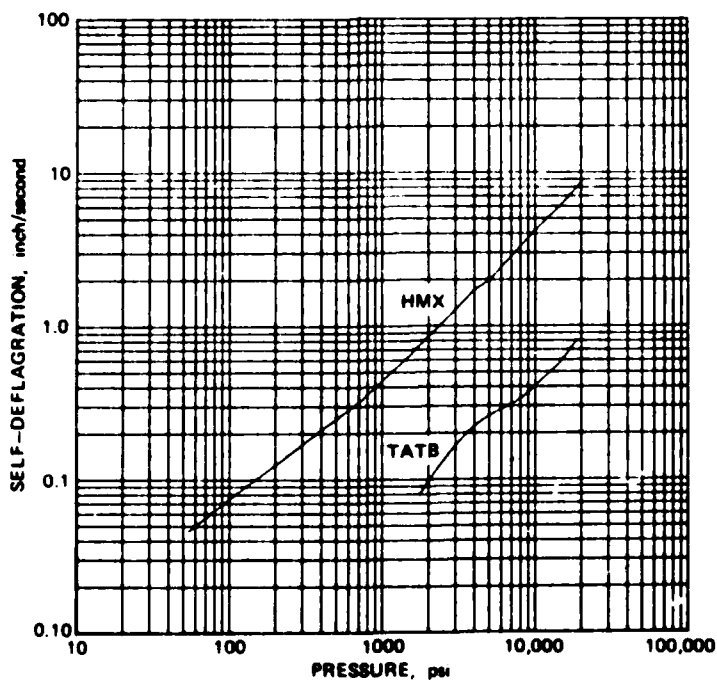


Figure 6. Burning Rates for HMX and TATB Pellets

Plans:

CFY

1. Complete HMX decomposition in oxygen.
2. Investigate HMX decomposition in nitrogen.
3. Start the analysis of decomposition products.
4. Investigate liquid HMX.
5. Measure burn rate of AP/HMX mixtures.
6. Finish survey contribution to AIAA progress series.

CFY+1

1. Study parameters which influence decomposition of nitramines.
2. Apply results in propellant combustion.
3. Study ignition of nitramine containing propellants.

CFY+2

1. Continue above.
2. Apply kinetic data in HMX deflagration.
3. Apply results to hazards problems.

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